DECLARATION

I, Ryuichi YAMADA, a Japanese Patent Attorney registered No. 7898 having my Business Office at Hasegawa Bldg., 4F, 7-7 Toranomon 3-chome, Minato-ku, Tokyo, Japan, solemnly and sincerely declare:

That I have a thorough knowledge of Japanese and English languages; and

That the attached pages contain a correct translation into English of the specification of the following Japanese Application:

APPLICATION NUMBER	DATE OF APPLICATION
255537/2001(Pat.)	27/AUG/2001

Applicant(s)

CANON KABUSHIKI KAISHA

Signed this 18th day of September, 2003.

Ryuichi YAMADA

PATENT OFFICE

JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

APPLICATION NUMBER	DATE OF APPLICATION
255537/2001(Pat.)	27/AUG/2001

Applicant(s)

CANON KABUSHIKI KAISHA

19/OCT/2001

Director-General,
Patent Office : KOZO OIKAWA (Seal)

Certificate No. 3092035/2001

<FILE NO.> 4529008

<DOCUMENT> PATENT APPLICATION
<FILE NO.> 4529008
<FILING DATE> 27/AUG/2001
<DIRECTED TO> The Director General of the Patent Office
<INTERNATIONAL CLASSIFICATION> H05B 33/00
<TITLE OF THE INVENTION>
LUMINESCENCE DEVICE AND DISPLAY APPARATUS
<NUMBER OF CLAIMS> 11

<INVENTOR(S)>
<Address> c/o CANON KABUSHIKI KAISHA
3-30-2 Shimomaruko, Ohta-ku, Tokyo
<Name> Takao TAKIGUCHI
<Address> Ditto
<Name> Akira TSUBOYAMA
<Address> Ditto
<Name> Shinjiro OKADA
<Address> Ditto
<Name> Satoshi IGAWA
<Address> Ditto
<Name> Jun KAMATANI
<Address> Ditto
<Name> Manabu FURUGORI
<Address> Ditto
<Name> Seishi MIURA
<Address> Ditto
<Name> Takashi MORIYAMA
<Address> Ditto
<Name> Hidemasa MIZUTANI

<APPLICANT>
<IDENTIFICATION NO.> 000001007
<NAME> CANON KABUSHIKI KAISHA
<ATTORNEY>
<IDENTIFICATION NO.> 100096828
<PATENT ATTORNEY>
<NAME> Keisuke WATANABE
<TEL> 03-3501-2138
<DESIGNATED ATTORNEY>
<IDENTIFICATION NO.> 100059410
<PATENT ATTORNEY>
<NAME> Yoshio TOYODA
<TEL> 03-3501-2138
<CLAIM FOR PRIORITY>
<APPLICATION NO.> 292492/2000
<FILING DATE> 26/SEP/2000
<CLAIM FOR PRIORITY>
<APPLICATION NO.> 358741/2000
<FILING DATE> 27/NOV/2000
<OFFICIAL FEES>
<PREPAY REGISTRATION NO.> 004938
<AMOUNT> 21000
<ARTICLES PRESENTED>
<ARTICLE> Specification 1
<ARTICLE> Drawing(s) 1
<ARTICLE> Abstract 1
<GENERAL POWER NO.> 9703710
<REQUEST FOR PROOF> YES

[Document]

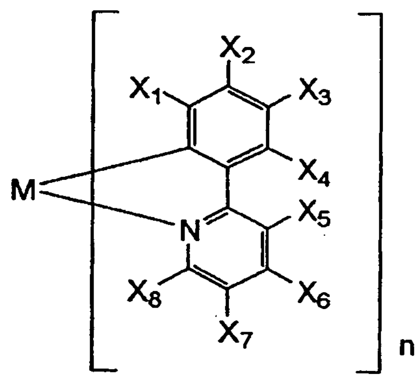
Specification

[Title of the Invention]

Luminescence Device and Display Apparatus

[Claims]

1. A luminescence device, comprising: an organic compound layer comprising a metal coordination compound represented by the following formula (1):



(1),

{wherein M denotes Ir, Rh or Pd; n is 2 or 3; and X1 to X8 independently denote hydrogen atom or a substituent [selected from the group consisting of halogen atom; nitro group; trifluoromethyl group trialkylsilyl group (which alkyl groups are independently a linear or branched alkyl group each having 1 - 8 carbon atoms); and a linear or branched alkyl group having 2 - 20 carbon atoms wherein one or at least two non-neighboring methylene groups which

can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and hydrogen atom in said alkyl group can be replaced with fluorine atom)]; with the proviso that at least one of X1 to X8 is a substituent other than hydrogen atom, and X2 and X3 cannot be fluorine atom at the same time}.

2. A device according to Claim 1, wherein in the formula (1), at least two of X1 to X8 are substituents other than hydrogen atom.

3. A device according to Claim 1 or 2, wherein in the formula (1), at least one of X5 to X8 is a substituent other than hydrogen atom.

4. A device according to any one of Claims 1 - 3, wherein in the formula (1), at least two of X1 to X4 are substituents other than hydrogen atom.

5. A device according to any one of Claims 1 - 4, wherein in the formula (1), at least one of X2, X3 and X4 has a Hammett's substituent constant of at least 0.2, and the metal coordination compound exhibits a peak emission wavelength at a normal temperature of at most 490 nm.

6. A device according to any one of Claims 1 -

5, wherein in the formula (1), X2, X3 and X4 provides a sum of Hammett's substituent constant of at least 0.41, and the metal coordination compound exhibits a peak emission wavelength at a normal temperature of at most 490 nm.

7. A device according to Claim 6, wherein the sum of Hammett's substituent constant is at least 0.50.

8. A luminescence device, comprising: an organic compound layer comprising a metal coordination compound which has a substituent having a Hammett's substituent constant of at least 0.2, and exhibits a peak emission wavelength at a normal temperature of at most 490 nm.

9. A device according to Claim 8, wherein said metal coordination compound has a plurality of substituents each having a Hammett's substituent constant of at least 0.2.

10. A device according to any one of Claims 1 - 9, further comprising two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the electrodes to produce

luminescence.

11. A display apparatus, comprising: a luminescence device according to any one of Claims 1 - 9 and a portion for driving the luminescence device.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Pertains]

The present invention relates to a luminescence device using an organic compound and a display apparatus. More specifically, the present invention relates to an organic electroluminescence device employing a metal coordination compound represented by the above-mentioned formula (1) as a luminescence material, and a display apparatus.

[0002]

[Prior Art]

An organic EL (electroluminescence) device has been extensively studied as a luminescence device with a high responsiveness and high efficiency. The organic EL device generally has a basic structure as shown in Figure 1(a), (b) (e.g., as described in "Macromol. Symp.", 125, pp. 1 - 48 (1997)).

[0003]

As shown in Figure 1, the EL device is generally constituted by disposing, on a transparent

substrate 15, a plurality of organic film layers between a transparent electrode 14 and a metal electrode 11.

In Figure 1(a), the organic layers includes a luminescence layer 12 and a hole transport layer 13.

As transparent electrode 14, a film of ITO (indium tin oxide) having a larger work function is used so as to ensure a good hole injection performance from the transparent electrode 14 into the hole transport layer 13. As the metal electrode 11, a layer of metal material such as aluminum, magnesium, alloys thereof, etc., having a smaller work function is used so as to ensure a good electron injection performance into the organic layer(s).

These electrodes may be formed in a thickness of 50 - 200 nm.

[0005]

The luminescence layer 12 may be formed of, e.g., aluminum quinolinol complex (representative example thereof may include Alq3 described hereinafter) having an electron transporting characteristic and a luminescent characteristic. The hole transport layer 13 may be formed of, e.g., triphenyldiamine derivative (representative example thereof may include α-NPD described hereinafter) having an electron donating characteristic.

[0006]

The above-described EL device exhibits an electrical rectification characteristic, so that when an electric field is applied between the metal electrode 11 as a cathode and the transparent electrode 14 as an anode, electrons are injected from the metal electrode 11 into the luminescence layer 12 and holes are injected from the transparent electrodes 15.

[0007]

The thus-injected holes and electrons are recombined within the luminescence layer 12 to produce excitons, thus producing luminescence. At that time, the hole transport layer 13 functions as an electron-blocking layer to increase a recombination efficiency at the boundary between the luminescence layer 12 and the hole transport layer 13, thus enhancing a luminescence efficiency.

[0008]

Further, in Figure 1(b), an electron transport layer 16 is disposed between the metal electrode 11 and the luminescence layer 12 shown in Figure 1(a), whereby an effective carrier blocking performance can be ensured by separating functions of luminescence, electron transport and hole transport, thus allowing efficient luminescence. The electron transport layer 16 may be formed of, e.g., oxadiazole derivatives.

[0009]

Heretofore, in ordinary organic EL devices, fluorescence produced during a transition of luminescent center molecule from a singlet excited state to a ground state has been used as luminescence. On the other hand, not the above fluorescence (luminescence) via singlet exciton, phosphorescence (luminescence) via triplet exciton has been studied for use in organic EL device as described in, e.g., Document 1: "Improved energy transfer in electrophosphorescent device" (D.F. O'Brien et al., Applied Physics Letters, Vol. 74, No. 3, pp. 442 - (1999)) and Document 2: "Very high-efficiency green organic light-emitting devices based on electrophosphorescence" (M.A. Baldo et al., Applied Physics Letters, Vol. 75, No. 1, pp. 4 - 1999)).

[0010]

In these documents, a four layer-structure of organic layers as shown in Figure 1(c) is principally used. The structure includes a hole transfer layer 13, a luminescence layer 12, an exciton diffusion-prevention layer 17, and an electron transport layer 16 successively formed in this order from the anode side. The materials used therefor are carrier transporting materials and phosphorescent materials shown below. Abbreviations for the respective materials are follows.

Alq3: aluminum-quinolinol complex,

α-NPD: N4,N4'-di-naphthalene-1-yl-N4,N4'-diphenyl-biphenyl-4,4'-diamine,

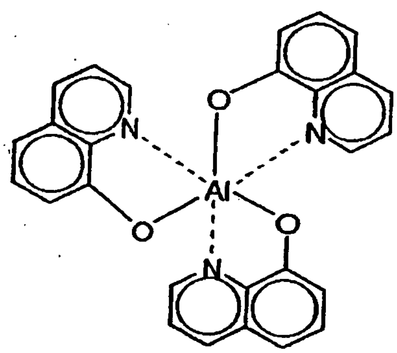
CBP: 4,4'-N,N'-dicarbazole-biphenyl,

BCP: 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline,

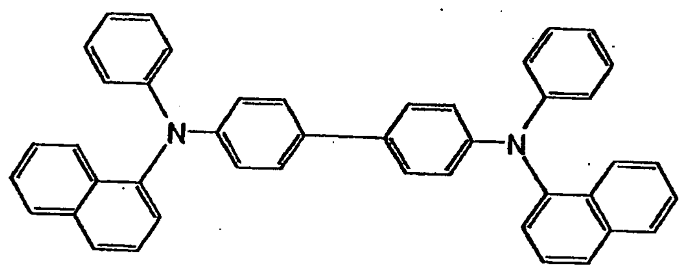
PtEOP: platinum-octaethyl porphine complex, and

Ir(ppy)$_3$: iridium-phenylpyridine complex.

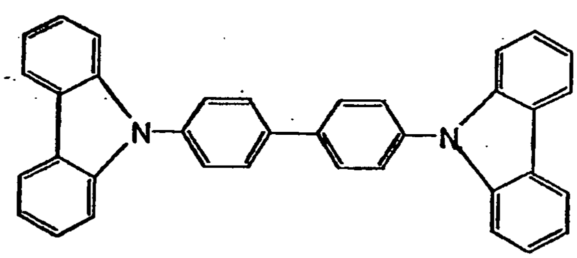
[0011]



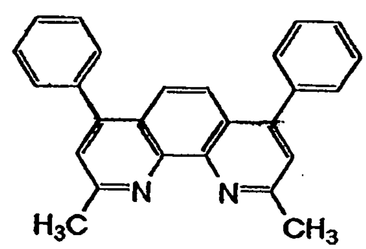
Alq3



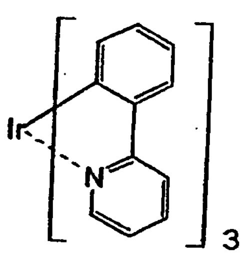
α -NPD



CBP



BCP



Ir(ppy)$_3$

[0012]

In the above Documents 1 and 2, higher efficiencies have been achieved by using a hole transport layer 13 of α-NPD, an electron transport layer 16 of Alq3, an exciton diffusion-prevention layer 17 of BPC, and a luminescence layer 12 of a mixture of CPB as a host material with $Ir(ppy)_3$ or PtOEP doped into CBP at a concentration of ca. 6 wt. %.

[0013]

The reason why the phosphorescence (luminescence) material has particularly attracted notice is that the phosphorescence material is expected to provide a higher luminescence efficiency in principle. The reason is as follows. Excitons produced by recombination of carriers comprise singlet excitons and triplet excitons presented in a ratio of 1:3. Fluorescence produced during the transition from the singlet excited state to the ground state is utilized as luminescence in the conventional organic EL devices. However, a resultant luminescence efficiency is 25 % (being upper limit) based on all the produced excitons in principle.

However, if phosphorescence produced during transition from the triplet excited state is employed,

a resultant luminescence efficiency is expected to be at least three times (that of the case of fluorescence) in principle. In addition thereto, if intersystem crossing from the singlet excited state which is a higher energy level to the triplet excited state is taken into consideration, the luminescence efficiency of phosphorescence can be expected to be 100 %, which is four times that of fluorescence, in principle.

[0014]

The use of phosphorescence based on transition from the triplet excited state has also been proposed in, e.g., Japanese Laid-Open Patent Application (JP-A) 11-329739 (organic EL device and production process thereof), JP-A 11-256148 (luminescent material and organic EL device using the same) and JP-A 8-319482 (organic electroluminescent device).

[0015]

An iridium-phenylpyrimidine complex, closer to the present invention, having a methyl substituent has been described in "Preprint for the 61-th Scientific Lecture of the Applied Physics Society of Japan", the third volume, P.1117, 6p-ZH-1 (2000).

[0016]

Further, an iridium-phenylpyrimidine complex, closer to the present invention, having 4-, 5-fluorine

substituents (herein, referred to as a "metal coordination compound A" has been described in "Polymer Preprints", 41(1), pp. 770 - 771 (2000).

[0017]

[Problems to be Solved by the Invention]

The above-mentioned organic EL devices utilizing phosphorescence have accompanied with a problem of luminescent deterioration particularly in an energized state. The reason for luminescent deterioration has not been clarified as yet but may be attributable to such a phenomenon that the life of triplet exciton is generally longer than that of singlet exciton by at least three digits, so that molecule is placed in a higher-energy state for a long period to cause reaction with ambient substance, formation of exciplex or excimer, change in minute molecular structure, structural change of ambient substance, etc.

[0018]

Anyway, the phosphorescence luminescence device is expected to provide a higher luminescence efficiency as described above, while the device is accompanied with the problem of luminescent deterioration in energized state.

[0019]

Accordingly, an object of the present invention is to provide a luminescence device and a

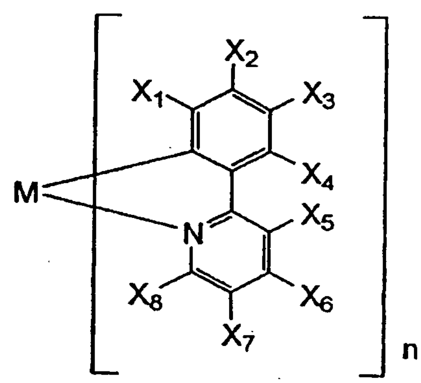
display apparatus, capable of providing a high-efficiency luminescence at a high brightness (or luminance) for a long period (less deterioration in luminescence in energized state).

[0020]

[Means for Solving the Problems]

More specifically, the present invention provided a luminescence device, comprising: an organic compound layer comprising a metal coordination compound represented by the following formula (1):

[0021]



(1),

[0022]

{wherein M denotes Ir, Rh or Pd; n is 2 or 3; and X1 to X8 independently denote hydrogen atom or a substituent [selected from the group consisting of halogen atom; nitro group; trifluoromethyl group trialkylsilyl group (which alkyl groups are independently a linear or branched alkyl group each having 1 - 8 carbon atoms); and a linear or branched alkyl group having 2 - 20 carbon atoms wherein one or

at least two non-neighboring methylene groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and hydrogen atom in said alkyl group can be replaced with fluorine atom)]; with the proviso that at least one of X1 to X8 is a substituent other than hydrogen atom, and X2 and X3 cannot be fluorine atom at the same time}.

[0023]

In the luminescence device of the present invention, in the formula (1), at least two of X1 to X8 may preferably be substituents other than hydrogen atom; at least one of X5 to X8 may preferably be a substituent other than hydrogen atom; and at least two of X1 to X4 may preferably be substituents other than hydrogen atom.

[0024]

Further, in the formula (1), it is preferable that at least one of X2, X3 and X4 has a Hammett's substituent constant of at least 0.2, and the metal coordination compound exhibits a peak emission wavelength at a normal temperature of at most 490 nm. In the formula (1), X2, X3 and X4 may preferably provide a sum of Hammett's substituent constant of at least 0.41, more preferably at least 0.50, and the metal coordination compound may preferably exhibit a peak emission wavelength at a normal temperature of at most 490 nm.

[0025]

The present invention provides a luminescence device, comprising: an organic compound layer comprising a metal coordination compound which has a substituent having a Hammett's substituent constant of at least 0.2, and exhibits a peak emission wavelength at a normal temperature of at most 490 nm. The metal coordination compound may preferably have a plurality of substituents each having a Hammett's substituent constant of at least 0.2.

[0026]

Further, the aforementioned luminescence device may preferably further comprise two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the electrodes to produce luminescence.

[0027]

Further, the present invention provide a display apparatus, comprising: the above-mentioned luminescence device and a portion for driving the luminescence device.

[0028]

[Embodiments for Practicing the Invention]

In the case where a luminescence layer is formed of a carrier transporting host material and a phosphorescent guest material, a process of emission

of light from the triplet exciton may generally involve the following steps:

(1) transport of electron and hole within a luminescence layer,

(2) formation of exciton of the host material,

(3) transmission of excited energy between host material molecules,

(4) transmission of excited energy from the host material molecule to the guest material molecule,

(5) formation of triplet exciton of the guest material, and

(6) phosphorescence produced during transition from the triplet excited state to the ground state of the guest material.

[0029]

In the respective steps, desired energy transmission and luminescence may be caused based on various deactivation and competition.

[0030]

In order to improve a luminescence efficiency of the EL device, a luminescence center material per se is required to provide a higher yield of luminescence quantum. However, an efficient energy transfer between host material molecules and/or between host material molecule and guest material molecule is also an important factor. Further, the case of the luminescent deterioration in energized

state has not been clarified as yet but may presumably relate to the luminescent center material per se or an environmental change thereof by its ambient molecular structure.

[0031]

For this reason, the present inventors have extensively studied on various metal coordination compounds and as a result, have found that the organic electroluminescence device using the metal coordination compound represented by the above-mentioned formula (1) as the luminescence center material allows a high-efficiency luminescence with a high brightness (luminance) for a long period and less luminescent deterioration in energized state.

[0032]

The metal coordination compound of formula (1) may preferably have substituents X1 to X8 in which at least two of X1 to X8 are substituents other than hydrogen atom. Further, in the formula (1), at least one of X5 to X8 may preferably be a substituent other than hydrogen atom and/or at least two of X1 to X4 may preferably be substituents other than hydrogen atom.

[0033]

The metal coordination compound used in the present invention has a phosphorescent property and is assumed to have a lowest excited state comprising, as a triplet excited state, a metal-to-

ligand charge transfer (MLCT*) state or π-π* excited state. The phosphorescent emission of light (phosphorescence) is produced during the transition from these states to the ground state.

[0034]

The luminescent material of the present invention provides a higher phosphorescence yield of 0.1 - 0.9 and a shorter phosphorescence life of 1 - 60 μsec.

More specifically, the shorter phosphorescence life is a condition for providing a resultant EL device with a higher luminescence efficiency. This is because the longer phosphorescence life increases molecules placed in their triplet excited state which is a waiting state for phosphorescence, thus resulting in a problem of lowering the resultant luminescence efficiency particularly at a higher current density. The material of the present invention is a suitable luminescent material for an EL device with a higher phosphorescence yield and a shorter phosphorescence life.

[0035]

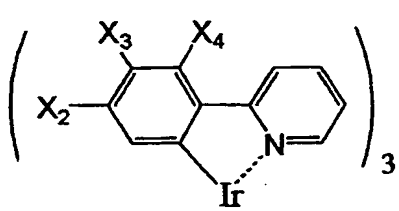
Further, it has been found that it is possible to control an emission wavelength of the metal coordination compound represented by the formula (1) by appropriately modifying the substituents X1 to

X8 thereof. In this regard, as a result of our investigation on various phosphorescence metal coordination compounds in order to search a blue luminescence material required to have a peak (maximum) emission wavelength of at most 490 nm, we have found that it is very effective to introduce a substituent having a Hammett's substituent constant of at least 0.2 into the metal coordination compound of formula (1) in order to provide a shorter peak emission wavelength.

[0036]

Hereinbelow, explanation will be made by taking the metal coordination compound represented by the formula (1) of the present invention as an example. We focus attention on a carbon atom connected to iridium metal of a metal coordination compound represented by the formula shown below, and have investigated a relationship between Hammett's substituent constants (σ) of substituents X2, X3 and X4, and peak emission wavelengths (λ_{PE}) of the metal coordination compound in toluene at room temperature, i.e., at 25 $^{\circ}C$.

[0037]



[0038]

A Hammett's substituent constant σm for meta-position was used for the substituents X2 and X4 and a Hammett's substituent constant σp for para-position was used for the substituent X3. When a plurality of substituents were present, a sum of the respective substituent constants (σm and σp) was used (as a Hammett's substituent constant σ). Here, Hammett's substituent constants σm and σp described on pages 96 - 103 (Table 1) of "Correlation between Structure and Activation of Drugs", Chemical Region Extra Edition 122, issued by Nanko-do (Japan) were used. A part of the substituent constants described therein is shown in Table 1 below.

[0039]

[Table 1]

Substituent	Hammett's σp	Constant σm
F	0.06	0.34
Cl	0.23	0.37
CF_3	0.54	0.43

[0040]

For example, when the Hammett's substituent constant σ of Example Compound No. (121) (X2 = F, X3 = CF_3) is calculated, 0.34 + 0.54 = 0.88. In a similar

manner, Hammett's substituent constants σ of Ex. Comp. Nos. (1), (32), (122) and (111) and the metal coordination compound A are calculated. The results are shown in Table 2 below together with corresponding peak (maximum) emission wavelength λ_{PE} in toluene. The results of Table 2 are plotted by taking the Hammett's substituent constant σ on the axis of abscissa and the peak emission wavelength λ_{PE} on the axis of ordinate (Figure 2).

[0041]

[Table 2]

Compound	σ	PE (nm)
Ex. Comp. No. (1)	0.06	522
Metal coordination compound A	0.40	505
Ex. Comp. No. (32)	0.54	487
Ex. Comp. No. (122)	0.68	471
Ex. Comp. No. (121)	0.88	466
Ex. Comp. No. (111)	0.91	479

[0042]

As apparent from Figure 2, introduction of substituent(s) having a larger Hammett's substituent constant is very effective to shorten the peak emission wavelength. Further, the metal coordination compound having the sum of peak emission wavelengths

of at least 0.41, particularly at least 0.50 is suitable as the blue luminescent material. A similar effect can be expected also for metal coordination compounds other than the metal coordination compound of the present invention. From also the viewpoints as described above, the metal coordination compound of the present invention is a suitable luminescent material for the EL device.

[0043]

Further, as shown in Examples appearing hereinafter, it has been clarified that the (metal coordination) compound of the present invention also has an excellent stability in a continuous energization test.

This may be attributable to introduction of particular substituents as a feature of the present invention allowing control of intermolecular interaction with a host luminescent material and suppression of formation of associated exciton leading to thermal inactivation, thus minimizing quenching to improve device characteristics. Further, the methyl group of methyl-substituted iridium-phenylpyrimidine complex described in the above-mentioned "Preprint for the 61-th Scientific Lecture of the Applied Physics Society of Japan", the third volume, P. 1117, 6p-ZH-1 (2000), has a smaller bulkiness than ethyl group and methoxy group in the present invention and a smaller

electronic effect than halogen atom, trifluoromethyl group and methoxy group in the present invention. As a result, the effect of controlling intermolecular interaction in the present invention cannot be expected.

[0044]

Further, compared with 4-, 5-fluorine substituted iridium-phenylpyrimidine complex described in "Proper Preprints", 2000, 41 (1), 770 - 771, it has been clarified that a luminescence device using the metal coordination compound according to the present invention has a higher durability as shown in Examples appearing later.

[0045]

In the case of phosphorescent (luminescent) material, luminescent characteristics are largely affected by its molecular environment. Principal characteristics of the fluorescent material are studied based on photoluminescence in the case of fluorescence device. However, results of photoluminescence (of the phosphorescent material) do not reflect luminescent characteristics of the resultant EL device in many cases since the luminescent characteristics in the case of the phosphorescence depend on a magnitude of polarity of ambient host material molecules, ambient temperature, whether presence state of the material is solid state

or liquid state, etc. Accordingly, it is generally difficult to expect the resultant EL characteristics (for the phosphorescent material) by simply removing a part of characteristics from photoluminescence results.

[0046]

The luminescence device according to the present invention, as shown in Figure 1, may preferably include the organic layer comprising the metal coordination compound between two oppositely disposed electrodes comprising a transparent electrode between which are supplied with a voltage to produce luminescence, thus constituting an electric-field luminescence device.

[0047]

The high-efficiency luminescence device shown in the present invention may be applicable to products required to allow energy saving and high luminance, applied examples of which may include those for display apparatus and illumination apparatus, a light source for printers, and backlight (unit) for a liquid crystal display apparatus. As the display apparatus, it is possible to provide a flat panel display apparatus capable of exhibiting an excellent energy saving performance, a high visibility and a good lightweight property. With respect to the light source for a printer, it becomes possible to replace a

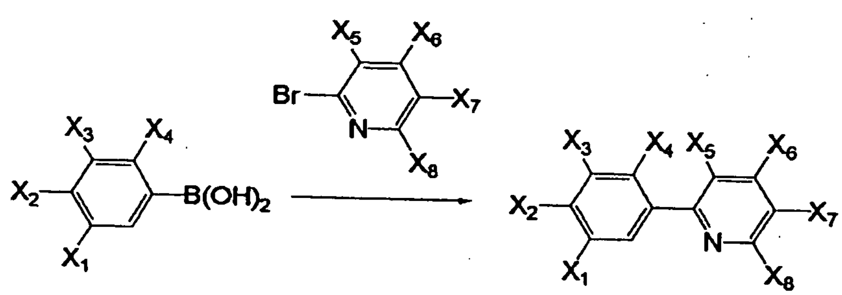
laser light source of laser beam printer currently used widely with the luminescence device according to the present invention. Further, when independently addressable devices are arranged in arrays and desired exposure of light to a photosensitive drum is effected to form an image. By the use of the device of the present invention, it becomes possible to considerably reducing the volume (size) of image forming apparatus. With respect to the illumination apparatus and backlight (unit), the resultant apparatus (unit) using the luminescence device of the present invention is expected to have an energy saving effect.

[0048]

A synthesis scheme of the metal coordination compound represented by the above-mentioned formula (1) used in the present invention will be shown by taking an iridium complex as an example.

Synthesis of ligand L

[0049]



:L

[0050]

Synthesis of iridium coordination compound

[0051]

$$Ir(CH_3COCHCOCH_3)_3 \xrightarrow{3 \times L} Ir(L)_3$$

[0052]

or

[0053]

$$IrCl_3.3H_2O \xrightarrow{2 \times L} [Ir(L)_2Cl]_2 \xrightarrow{L} Ir(L)_3$$

[0054]

Hereinafter, specific structural formulas of the metal coordination compound used in the present invention are shown below. However, these are merely exemplified as representative examples. The present invention is not restricted to these examples.

[0055]

[Table 3]

No.	M	n	X_1	X_2	X_3	X_4	X_5
(1)	Ir	3	H	H	F	H	H
(2)	Ir	3	H	F	H	H	H
(3)	Ir	3	H	H	Cl	H	H
(4)	Ir	3	H	H	F	H	H
(5)	Ir	3	H	H	F	H	H
(6)	Ir	3	H	C_2H_5	H	H	H
(7)	Ir	3	H	H	NO_2	H	H
(8)	Ir	3	H	H	NO_2	H	H
(9)	Ir	3	H	H	NO_2	H	H
(10)	Ir	3	H	H	NO_2	H	H
(11)	Ir	3	H	H	C_3H_7	H	H
(12)	Ir	3	H	C_2H_5	OCH_3	H	H
(13)	Ir	3	H	H	C_3H_7	H	H
(14)	Ir	3	H	$C_{20}H_{41}$	H	H	H
(15)	Ir	3	H	H	OCH_3	H	H
(16)	Ir	3	H	OCH_3	OCH_3	H	H
(17)	Ir	3	H	H	$OCH(CH_3)_2$	H	H
(18)	Ir	3	H	H	OC_5H_{11}	H	H
(19)	Ir	3	H	H	$OC_{16}H_{33}$	H	H
(20)	Ir	3	H	H	OCH_3	H	H
(21)	Ir	3	H	H	$OCH(CH_3)_2$	H	H
(22)	Ir	3	H	H	$OC_{10}H_{21}$	H	H
(23)	Ir	3	H	H	$OCH(CH_3)_2$	H	H
(24)	Ir	3	H	H	SCH_3	H	H
(25)	Ir	3	H	$OCH_2CH{=}CH_2$	H	H	H
(26)	Ir	3	H	H	$OCH_2C{\equiv}CCH_3$	H	H
(27)	Ir	3	H	H	$COCH_3$	H	H
(28)	Ir	3	H	H	$COCH_3$	H	H
(29)	Ir	3	H	H	$COCH_3$	H	H
(30)	Ir	3	H	H	$COCH_3$	H	H

[0056]

[Table 4]

No.	M	n	X_1	X_2	X_3	X_4	X_5
(31)	Ir	3	H	H	COC_9H_{19}	H	H
(32)	Ir	3	H	H	CF_3	H	H
(33)	Ir	3	H	H	CF_3	H	H
(34)	Ir	3	H	H	CF_3	H	H
(35)	Ir	3	H	H	CF_3	H	H
(36)	Ir	3	H	C_3F_7	H	H	H
(37)	Ir	3	H	H	OCF_3	H	H
(38)	Ir	3	H	OCF_3	H	H	H
(39)	Ir	3	H	H	OCF_3	H	H
(40)	Ir	3	H	H	OCF_3	H	H
(41)	Ir	3	H	H	OCF_3	H	H
(42)	Ir	3	H	H	$OCH_2C_3F_7$	H	H
(43)	Ir	3	H	$O(CH_2)_3C_2F_5$	H	H	H
(44)	Ir	3	H	H	$O(CH_2)_3OCH_2C_2F_5$	H	H
(45)	Ir	3	H	H	$COOC_2H_5$	H	H
(46)	Ir	3	H	$OCOCH_3$	H	H	H
(47)	Ir	3	H	H	$O(CH_2)_2C_3F_7$	H	H
(48)	Ir	3	H	H	H	H	H
(49)	Ir	3	H	H	H	H	H
(50)	Ir	3	H	H	H	H	H
(51)	Ir	3	H	H	$Si(CH_3)_3$	H	H
(52)	Ir	3	H	H	$Si(CH_3)_2C_4H_9$	H	H
(53)	Ir	3	H	$Si(CH_3)_2C_8H_{17}$	H	H	H
(54)	Ir	3	H	H	$Si(C_2H_5)_3$	H	H
(55)	Ir	3	H	H	H	H	H
(56)	Ir	3	H	C_2H_5	OCH_3	H	H
(57)	Ir	3	H	F	H	F	H
(58)	Ir	3	H	F	H	F	H
(59)	Ir	3	H	H	$Si(CH_3)_3$	H	H
(60)	Ir	3	H	$Si(CH_3)_2C_7H_{15}$	OCH_3	H	H

[0057]

[Table 5]

No.	M	n	X_1	X_2	X_3	X_4	X_5	X_6
(61)	Rh	3	H	H	F	H	H	H
(62)	Rh	3	F	F	H	H	H	H
(63)	Rh	3	H	H	F	H	H	OCH_3
(64)	Rh	3	H	H	NO_2	H	H	H
(65)	Rh	3	H	H	NO_2	H	H	OC_8H_{17}
(66)	Rh	3	H	H	C_2H_5	H	H	H
(67)	Rh	3	H	C_2H_5	OCH_3	H	H	H
(68)	Rh	3	H	$C_{12}H_{25}$	H	H	H	H
(69)	Rh	3	H	C_3H_7	H	H	H	OCH_3
(70)	Rh	3	H	H	$OCH(CH_3)_2$	H	H	H
(71)	Rh	3	H	H	$OC_{15}H_{31}$	H	H	H
(72)	Rh	3	H	H	OC_6H_{13}	H	H	NO_2
(73)	Rh	3	H	H	OCH_3	H	H	OCH_3
(74)	Rh	3	H	H	$OCH(CH_3)_2$	H	H	H
(75)	Rh	3	H	H	$OCH_2CH{=}CH_2$	H	H	H
(76)	Rh	3	H	$OC{\equiv}CC_4H_9$	H	H	H	H
(77)	Rh	3	H	H	SC_2H_5	H	H	H
(78)	Rh	3	H	H	SCH_3	H	H	OCH_3
(79)	Rh	3	H	SCH_3	SCH_3	H	H	H
(80)	Rh	3	H	H	$COCH_3$	H	H	H
(81)	Rh	3	H	H	$COCH_3$	H	H	OCH_3
(82)	Rh	3	H	H	CF_3	H	H	H
(83)	Rh	3	H	H	CF_3	H	H	$OCH(CH_3)_2$
(84)	Rh	3	H	H	OCF_3	H	H	H
(85)	Rh	3	H	H	$OCH_2C_4F_9$	H	H	H
(86)	Rh	3	H	H	$O(CH_2)_6C_2F_5$	H	H	H
(87)	Rh	3	H	H	H	H	H	OCH_3
(88)	Rh	3	H	H	$Si(CH_3)_3$	H	H	H
(89)	Rh	3	H	$Si(CH_3)_2C_6H_{13}$	H	H	H	H
(90)	Rh	3	H	$Si(CH_3)_2C_7H_{15}$	OCH_3	H	H	H

[0058]

[Table 6]

No.	M	n	X_1	X_2	X_3	X_4	X_5	X_6
(91)	Pd	2	H	H	F	H	H	H
(92)	Pd	2	H	F	H	F	H	H
(93)	Pd	2	H	H	F	H	H	OC_7H_{15}
(94)	Pd	2	H	H	NO_2	H	H	H
(95)	Pd	2	H	H	NO_2	H	H	OC_5H_{11}
(96)	Pd	2	H	C_2H_5	OCH_3	H	H	H
(97)	Pd	2	H	H	C_5H_{11}	H	H	OCH_3
(98)	Pd	2	H	$C_{15}H_{31}$	H	H	H	H
(99)	Pd	2	H	H	$OCH(CH_3)_2$	H	H	H
(100)	Pd	2	H	H	OC_3H_7	H	H	H
(101)	Pd	2	H	H	COC_8H_{17}	H	H	H
(102)	Pd	2	H	H	CF_3	H	H	H
(103)	Pd	2	H	H	CF_3	H	H	$OCH(CH_3)_2$
(104)	Pd	2	H	H	OCF_3	H	H	H
(105)	Pd	2	H	H	$Si(CH_3)_3$	H	H	H
(106)	Pd	2	H	H	F	H	H	OC_5H_{11}
(107)	Pd	2	H	H	NO_2	H	H	OC_3H_7
(108)	Pd	2	H	H	C_2H_5	H	H	OCH_3
(109)	Pd	2	H	$C_{10}H_{21}$	H	H	H	H
(110)	Pd	2	H	H	$COCH_3$	H	H	H
(111)	Ir	3	H	Cl	CF_3	H	H	H
(112)	Ir	3	H	Cl	CF_3	H	H	H
(113)	Ir	3	H	Cl	CF_3	H	H	OCH3
(114)	Rh	3	H	Cl	CF_3	H	H	H
(115)	Rh	3	H	Cl	CF_3	H	H	H
(116)	Rh	3	H	Cl	CF_3	H	H	CF3
(117)	Rh	3	H	Cl	CF_3	H	H	OCH3
(118)	Rh	3	H	Cl	CF_3	H	H	CH3
(119)	Pd	2	H	Cl	CF_3	H	H	H
(120)	Pd	2	H	Cl	CF_3	H	H	H

[0059]

[Table 7]

No.	M	n	X_1	X_2	X_3	X_4	X_5
(121)	Ir	3	H	F	CF_3	H	H
(122)	Ir	3	H	F	H	F	H
(123)	Ir	3	H	CF_3	H	CF_3	H
(124)	Ir	3	H	CF_3	H	F	H
(125)	Ir	3	H	CF_3	CF_3	H	H
(126)	Ir	3	F	C_2H_5	H	H	H
(127)	Ir	3	F	H	NO_2	H	H
(128)	Ir	3	F	H	NO_2	F	H
(129)	Ir	3	F	H	NO_2	H	H
(130)	Ir	3	F	H	NO_2	H	H
(131)	Ir	3	F	H	C_3H_7	H	H
(132)	Ir	3	F	C_2H_5	OCH_3	H	H
(133)	Ir	4	F	H	C_3H_7	H	H
(134)	Ir	3	H	$C_{20}H_{41}$	H	F	H
(135)	Ir	3	H	H	OCH_3	F	H
(136)	Ir	3	H	OCH_3	OCH_3	F	H
(137)	Ir	3	H	H	$OCH(CH_3)_2$	F	H
(138)	Ir	3	H	H	OC_5H_{11}	F	H
(139)	Ir	3	H	H	$OC_{16}H_{33}$	F	H
(140)	Ir	3	H	H	OCH_3	F	H
(141)	Ir	3	H	H	$OCH(CH_3)_2$	H	F
(142)	Ir	3	H	H	$OC_{10}H_{21}$	H	F
(143)	Ir	3	H	H	$OCH(CH_3)_2$	H	F
(144)	Ir	3	H	H	SCH_3	H	CH_3
(145)	Ir	3	H	$OCH_2CH{=}CH_2$	H	H	CH_3
(146)	Ir	3	H	H	$OCH_2C{\equiv}CCH_3$	H	H
(147)	Ir	3	H	H	$COCH_3$	H	H
(148)	Ir	3	H	H	$COCH_3$	H	H
(149)	Ir	3	H	H	$COCH_3$	H	H
(150)	Ir	3	CF_3	H	$COCH_3$	H	H

[0060]

[Table 8]

No.	M	n	X_1	X_2	X_3	X_4	X_5
(151)	Ir	3	F	H	COC_9H_{19}	H	H
(152)	Ir	3	H	CF_3	H	F	H
(153)	Ir	3	F	H	CF_3	H	H
(154)	Ir	3	H	H	CF_3	F	H
(155)	Ir	3	H	H	CF_3	F	H
(156)	Ir	3	H	C_3F_7	H	CF_3	H
(157)	Ir	3	H	H	OCF_3	H	CF_3
(158)	Ir	3	H	OCF_3	H	H	CH_3
(159)	Ir	3	H	CF_3	H	CF_3	H
(160)	Ir	3	H	H	OCF_3	H	F
(161)	Ir	3	H	H	OCF_3	H	H
(162)	Ir	3	H	H	$OCH_2C_3F_7$	H	H
(163)	Ir	3	H	$O(CH_2)_3C_2F_5$	H	H	H
(164)	Ir	3	H	H	$O(CH_2)_3OCH_2C_2F_5$	Cl	H
(165)	Ir	3	H	H	$COOC_2H_5$	F	H
(166)	Rh	3	H	$OCOCH_3$	H	H	F
(167)	Rh	3	H	H	$O(CH_2)_2C_3F_7$	H	CH_3
(168)	Rh	3	H	H	H	H	H
(169)	Rh	3	H	H	H	H	H
(170)	Rh	3	H	H	H	H	H
(171)	Rh	3	H	H	$Si(CH_3)_3$	H	H
(172)	Rh	3	H	H	$Si(CH_3)_2C_4H_9$	H	H
(173)	Rh	3	H	$Si(CH_3)_2C_8H_{17}$	H	F	H
(174)	Rh	3	H	H	$Si(C_2H_5)_3$	F	H
(175)	Rh	3	H	H	H	F	H
(176)	Pd	2	H	C_2H_5	OCH_3	F	H
(177)	Pd	2	F	H	F	F	H
(178)	Pd	2	F	H	F	F	H
(179)	Pd	2	F	H	$Si(CH_3)_3$	H	H
(180)	Pd	2	F	$Si(CH_3)_2C_7H_{15}$	OCH_3	H	H

[0061]

<Examples>

Examples 1-10 and Comparative Example 1

A common portion of device preparation steps used in the present invention will be described.

[0062]

As a device structure, a device having a three-layer structure of organic layers shown in (b) of Figure 1 was used. On a glass substrate (transparent substrate 15), a 100 nm-thick film (transparent electrode 14) of ITO (indium tin oxide) was formed by sputtering, followed by patterning to have an (opposing) electrode area of 3 mm^2. On the ITO-formed substrate, organic layers and metal electrode layers shown below were successively formed in film by vacuum (vapor) deposition using resistance heating in a vacuum chamber (10^{-4} Pa).

Organic layer 1 (hole transport layer 13) (40 nm): α-NPD

Organic layer 2 (luminescence layer 12) (30 nm): mixture of CBP:metal coordination compound (metal coordination compound weight proportion: 5 wt. %)

Organic layer 3 (electron transport layer 16) (30 nm): Alq3

Metal electrode layer 1 (15 nm): Al-Li alloy (Li = 1.8 wt. %)

Metal electrode layer 2 (100 nm): Al

[0063]

An electric field was applied between the ITO electrode (as an anode) and the Al electrode (as a cathode) to apply a voltage to each device so as to provide the device with the same current value, thus measuring a luminance (brightness) with time. The constant current amount was 70 mA/cm^2. At that time, the resultant luminances of the respective devices were in the range of 70 - 210 cd/m^2.

[0064]

As the cause of device deterioration, oxygen or moisture (water content) is a problematic factor, so that each device was subjected to the above measurement in a dry nitrogen gas stream after it was taken out of the vacuum chamber so as to remove such a factor.

[0065]

In Comparative Example 1, as the conventional luminescent material, $Ir(ppy)_3$ described in the aforementioned document 2.

[0066]

[Table 9]

Ex. No.	Luminescence material No.	Luminance half-life (Hr)
1	(4)	750
2	(7)	500
3	(17)	900
4	(18)	850
5	(21)	850
6	(23)	500
7	(32)	600
8	(56)	700
9	(67)	400
10	(74)	450
Comp.Ex. 1	$Ir(ppy)_3$	350

[0067]

The results of energization durability test of the devices using the respective compounds are shown in Table 9. The devices (of the present invention) provide luminance half-life times clearly longer than the device using the conventional luminescent material, so that it becomes possible to realize a high-durability device resulting from stability of the materials of the present invention.

[0068]

Examples 11 - 13 and Comparative Example 2

In these examples, a device having a four-layer structure of organic layers shown in (c) of Figure 1 was used.

On a glass substrate (transparent substrate 15), a 100 nm-thick film (transparent electrode 14) of ITO (indium tin oxide) was formed, followed by patterning to have an (opposing) electrode area of 3 mm^2. On the ITO-formed substrate, organic layers and metal electrode layers shown below were successively formed in film by vacuum (vapor) deposition using resistance heating in a vacuum chamber (10^{-4} Pa).

Organic layer 1 (hole transport layer 13) (40 nm): α-NPD

Organic layer 2 (luminescence layer 12) (20 nm): mixture of CBP: predetermined coordination compound (weight proportion: 7 wt. %)

Organic layer 3 (exciton diffusion prevention layer 17) (10 nm): BCP

Organic layer 4 (electron transport layer 16) (30 nm): Alq3

Metal electrode layer 1 (15 nm): Al-Li alloy (Li = 1.8 wt. %)

Metal electrode layer 2 (100 nm): Al

[0069]

As the metal coordination compounds, Ex. Comp. Nos. (1), (32) and (49) were used. These

compounds were subjected to measurement of photoluminescence spectrum in order to evaluate a luminescent characteristic of these metal coordination compounds. Each of the metal coordination compounds in a solution state in toluene at a concentration of 10^{-4} mol/l and at 25 ^{o}C was subjected to measurement of photoluminescence spectrum by using a spectrophoto-fluorometer ("Model F4500", mfd. by Hitachi K.K.). An excited light of approximately 350 nm was used. The values of photoluminescence spectrum of the metal coordination compounds were substantially equivalent to those in the EL devices under voltage application, so that it was confirmed that luminescence produced by the EL device was based on luminescence from the metal coordination compound used.

[0070]

Experimental results of the above photoluminescence and EL devices were shown in Table 10.

[0071]

Characteristics of the EL devices were measured by using a microammeter ("Model 4140B", mfd. by Hewlett-Packard Co.) for current-voltage characteristic, using a spectrophotofluoro-meter ("Model SR1", mfd. by Topcon K.K.) for luminescence spectrum, and using a luminance meter ("Model BM7",

mfd. by Topcon K.K.) for a luminescence luminance.

In these examples, the respective devices corresponding to the respective coordination compounds showed a good electrical rectification characteristic.

[0072]

Although the luminescence device using $Ir(ppy)_3$ (Comparative Example 2) shows a maximum (peak emission wavelength) of emission spectrum (λ_{PE}) of 510 nm, the EL devices using the metal coordination compounds used in the present invention are found to show longer peak emission wavelengths than $Ir(ppy)_3$ by approximately 40 - 110 nm, thus resulting in smaller relative luminous efficiencies.

Although smaller energy conversion efficiencies and luminescence efficiencies of the luminescence devices of the present invention are obtained compared with those of the luminescence device using $Ir(ppy)_3$, this may be attributable to the smaller relative luminous efficiencies with the longer peak emission wavelengths, thus not attributable to essentially inferior luminescent characteristics of the luminescence devices using the metal coordination compounds of these Examples.

[0073]

With respect to the luminance half-lifes of the luminescence devices, experiment was performed in the same manner as in Examples 1 - 11. Compared with

the luminescence device using $Ir(ppy)_3$, the luminescence devices using the metal coordination compounds of these Examples are found to show longer luminance half-lifes.

[0074]

[Table 10]

Ex.No	Luminescent material No. (Ex.Comp.No.)	λ PE in toluene (nm)	λ PE (nm)	Energy conversion efficiency (lm/W)	Luminescence efficiency (cd/A)	Current density (mA/cm^2 at 12V)	Luminance half-life (Hr)
Comp. Ex. 2	$Ir(ppy)_3$	510	510	6.0	19.0	20	150
11	(1)	522	525	4.0	13.6	170	300
12	(32)	487	525	0.4	2.4	130	400
13	(49)	537	545	2.1	7.0	25	250

[0075]

Synthesis examples of the metal coordination compounds used in the present invention will be shown below.

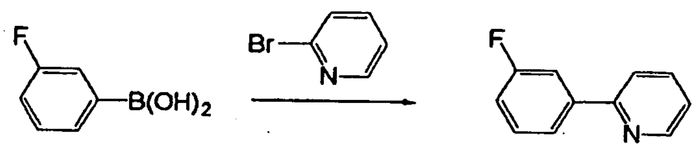
[0076]

<u>Example 14</u> (Synthesis of Ex. Comp. No. (1))

In a 1 liter-three necked flask, 20.0 g (126.6 mM) of 2-bromopyridine, 17.7 g (126.4 mM) of 3-fluorophenylbronic acid, 130 ml of toluene, 65 ml of ethanol and 130 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 4.60 g (3.98 mM) of tetrakis (triphenyl-phosphine) palladium (0) was added, followed by heat-refluxing for 6 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/ethyl acetate = 5/1) to obtain 6.0 g of 2-(3-fluorophenyl)pyridine (pale brown liquid) (Yield: 34.6 %).

[0077]

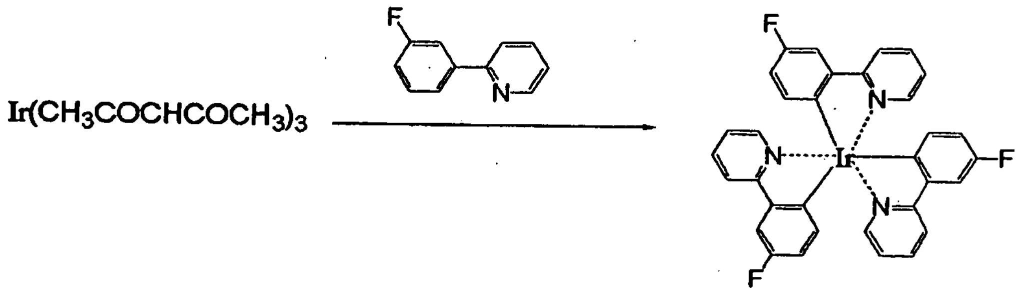


[0078]

In a 100 ml-four necked flask, 50 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 °C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 1.04 g (6.00 mM) of 2-(3-fluorophenyl)pyridine and 0.50 g (1.02 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 10 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 300 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 5 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) to obtain 0.22 g of Iridium (III) tris[2-(3-fluorophenyl)pyridine] (yellow powder) (Yield: 31.0 %).

[0079]



[0080]

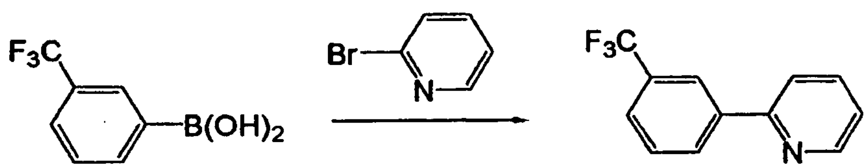
<u>Example 15</u> (Synthesis of Ex. Comp. No. (32))

In a 1 liter-three necked flask, 20.8 g (131.6 mM) of 2-bromopyridine, 25.0 g (131.6 mM) of 3-trifluoromethylphenylbronic acid, 130 ml of toluene, 65 ml of ethanol and 130 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 4.76 g (4.12 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 7 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue (pale brown liquid). The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 1/1) to obtain 6.0 g of 2-(3-trifluoromethylphenyl)pyridine (pale brown liquid)

(Yield: 21.1%).

[0081]



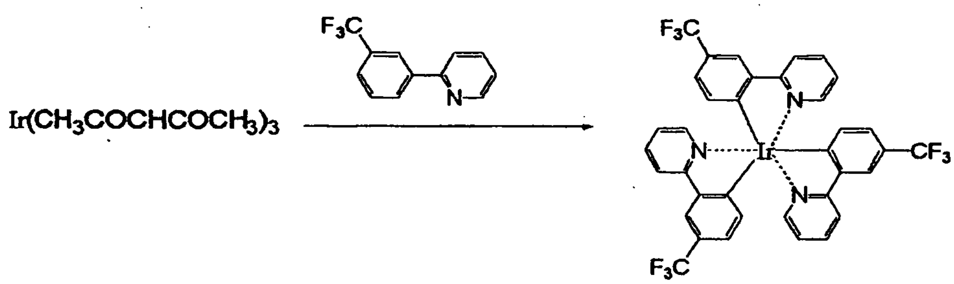
[0082]

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{o}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{o}C by standing. To glycerol, 2.68 g (12.0 mM) of 2-(3-trifluoromethylphenyl)pyridine and 1.00 g (2.04 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 10 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 5 hours at 100 ^{o}C under reduced pressure. The precipitate was dissolved in chloroform and the insoluble matter was removed by filtration, followed by purification by silica gel column chromatography (eluent: chloroform) and recrystallization from a

mixture solvent (chloroform/methanol) to obtain 0.62 g of Iridium (III) tris[2-(3-trifluoromethylphenyl)-pyridine] (yellow powder) (Yield: 35.3 %) showed a peak emission wavelength max of emission spectrum in toluene at 25 °C of 487 nm.

[0083]



[0084]

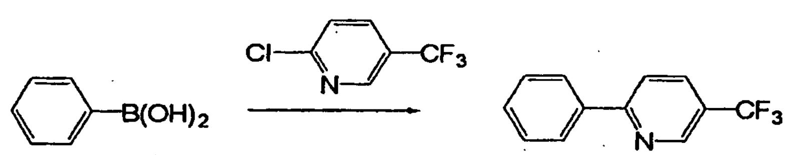
<u>Example 16</u> (Synthesis of Ex. Comp. No. (49))

In a 1 liter-three necked flask, 25.6 g (141.0 mM) of 2-chloro-5-trifluoromethylpyridine, 17.2 g (141.0 mM) of phenylbronic acid, 140 ml of toluene, 70 ml of ethanol and 140 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 5.10 g (4.41 mM) of tetrakis (triphenyl-phosphine) palladium (0) was added, followed by heat-refluxing for 6 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and

toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 5/1). The resultant creamy crystal was purified by alumina column chromatography (eluent: toluene) and recrystallized from ethanol to obtain 13.1 g of 2-phenyl-5-trifluoromethylpyridine (colorless crystal) (Yield: 41.6 %).

[0085]



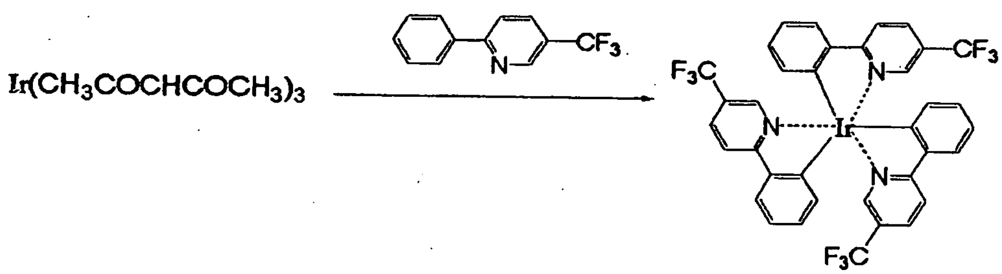
[0086]

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 °C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 2.68 g (12.0 mM) of 2-phenyl-5-trifluoromethylpyridine and 1.00 g (2.04 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of

1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 4 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) to obtain 0.43 g of Iridium (III) tris-(2-phenyl-5-trifluoromethylpyridine) (orange powder) (Yield: 24.5 %).

[0087]



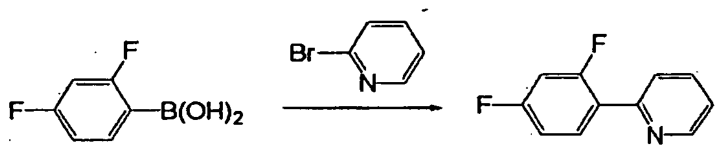
[0088]

Example 17 (Synthesis of Ex. Comp. No. (122))

In a 100 ml-three necked flask, 3.16 g (19.9 mM) of 2-bromopyridine, 3.16 g (20.0 mM) of 2,4-difluorophenylbronic acid, 15 ml of toluene, 7.5 ml of ethanol and 15 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 0.72 g (0.62 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 8 hours and 40 minutes under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and ethyl acetate. The organic layer was washed with water followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/ethyl acetate = 10/1) to obtain 3.28 g of 2-(2,4-difluorophenyl)pyridine (pale yellow oily product) (Yield: 86.0 %).

[0089]



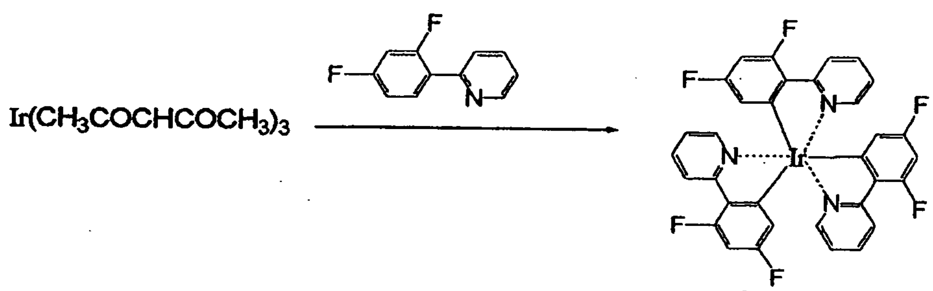
[0090]

In a 100 ml-four necked flask, 50 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 °C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 0.96 g (5.02 mM) of 2-(2,4-difluorophenyl)pyridine and 0.50 g (1.02 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 10 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 300 ml of 1N-HCl. The resultant precipitate was recovered by

filtration and washed with water, followed by drying for 5 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) and recrystallization from a mixture solvent (chloroform/methanol) to obtain 0.25 g of Iridium (III) tris[2-(4,6-difluorophenyl)-pyridine] (yellow powder) (Yield: 32.1 %), which showed a peak emission wavelength λmax of emission spectrum in toluene at 25 °C of 471 nm.

[0091]



[0092]

<u>Example 18</u> (Synthesis of Ex. Comp. No. (121))

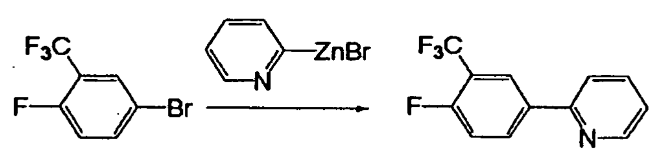
In a 500 ml-three necked flask, 11.0 g (45.3 mM) of 5-bromo-2-fluorobenzotrifluoride and 90 ml of dry tetrahydrofuran (THF) were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 2.60 g (2.25 mM) of tetrakis(triphenylphosphine) palladium (0) was added, followed by cooling to 20 - 21 °C (inner temperature) on an ice bath in nitrogen gas stream. At that

temperature, 90 ml of 0.5 M-THF solution of 2-pyridylzinc bromide was gradually added dropwise to the mixture in nitrogen gas stream, followed by stirring for 4 hours at that temperature.

After the reaction, the reaction mixture was poured into cool water, followed by addition of ethyl acetate to remove the insoluble matter by filtration. The organic layer was washed with water and dried with anhydrous sodium sulfate, followed by distilling-off of the solvent under reduced pressure to obtain a residue.

The residue was purified by silica gel column chromatography (eluent: hexane/ethyl acetate = 20/1) to obtain 1.80 g of 2-(4-fluoro-3-trifluoromethyl-phenyl)pyridine (pale brown oily product) (Yield: 16.6 %).

[0093]



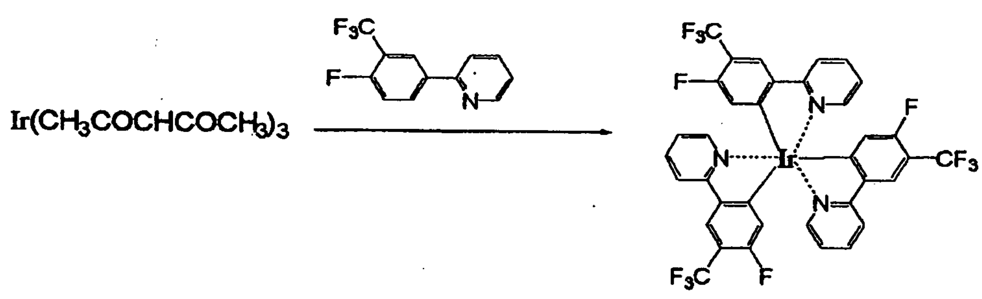
[0094]

In a 100 ml-four necked flask, 50 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{O}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{O}C by standing. To glycerol, 1.21 g (5.02 mM) of 2-(4-

fluoro-3-trifluoromethylphenyl)pyridine and 0.50 g (1.02 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 10 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 300 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 5 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) and recrystallization from a mixture solvent (chloroform/methanol) to obtain 0.20 g of Iridium (III) tris[2-(4-fluoro-5-trifluoromethyl-phenyl)pyridine] (yellow powder) (Yield: 21.5 %), which showed a peak emission wavelength λmax of emission spectrum in toluene at 25 °C of 466 nm.

[0095]



[00096]

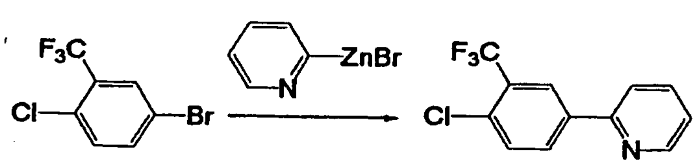
<u>Example 19</u> (Synthesis of Ex. Comp. No. (111))

In a 500 ml-three necked flask, 11.8 g (45.5 mM) of 5-bromo-2-chlorobenzotrifluoride and 90 ml of

dry tetrahydrofuran (THF) were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 2.60 g (2.25 mM) of tetrakis(triphenylphosphine) palladium (0) was added, followed by cooling to 13.5 - 14 ^{O}C (inner temperature) on an ice bath in nitrogen gas stream. At that temperature, 90 ml of 0.5 M-THF solution of 2-pyridylzinc bromide was gradually added dropwise to the mixture in nitrogen gas stream, followed by stirring for 3 hours at ca. 20 ^{O}C.

After the reaction, the reaction mixture was poured into cool water, followed by addition of ethyl acetate to remove the insoluble matter by filtration. The organic layer was washed with water and dried with anhydrous sodium sulfate, followed by distilling-off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: hexane/ethyl acetate = 10/1) to obtain 3.70 g of 2-(4-chloro-5-trifluoromethylphenyl)pyridine (pale brown oily product) (Yield: 31.9 %).

[0097]

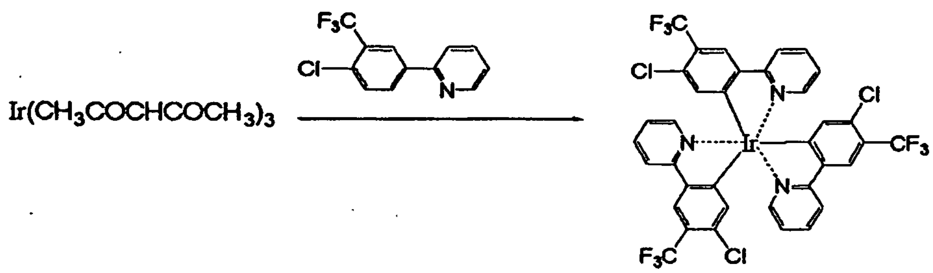


[0098]

In a 100 ml-four necked flask, 50 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{o}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{o}C by standing. To glycerol, 1.29 g (5.01 mM) of 2-(4-chloro-3-trifluoromethylphenyl)pyridine and 0.50 g (1.02 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 300 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 5 hours at 100 ^{o}C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) and recrystallization from a mixture solvent (chloroform/hexane) to obtain 0.25 g of Iridium (III) tris[2-(4-chloro-3-trifluoromethylphenyl)pyridine] (yellow powder) (Yield: 25.4 %), which showed a peak emission wavelength λmax of emission spectrum in toluene at 25 ^{o}C of 479 nm.

[0099]



[0100]

Comparative Example 3 (Synthesis of metal coordination compound A)

Iridium (III) tris[2-(4,5-difluoromethylphenyl)pyridine metal coordination compound A described in Polymer Preprints, 41(1), pp. 770 - 771 (2000) was prepared in the same manner as in Example 17 except that 2,4-difluorophenylbronic acid was changed to 3,4-difluorophenylbronic acid.

The metal coordination compound A showed a peak emission wavelength λmax of emission spectrum in toluene at 25 ^{o}C of 505 nm.

[0101]

Example 20 and Comparative Example 4

Two luminescence devices were prepared and subjected to continuous energization test in the same manner as in Example 1 by using the metal coordination compound (Ex. Comp. No. (122)) and the metal coordination compound A prepared in Comparative Example 3, respectively.

The results are shown in Table 11 below.

[0102]

[Table 11]

Ex. No.	Luminescent material No. (Ex. Comp. No.)	Luminance half-life (Hr)
20	(122)	630
Comp.Ex. 4	Metal coordination compound A	310

[0103]

As apparent from Table 18, the luminescence device using the metal coordination compound of formula (1) according to the present invention exhibited a luminance half-life considerably longer than that of the luminescence device using the metal coordination compound A, thus resulting in an EL device excellent in durability (luminance stability).

[0104]

[Effect of the Invention]

As described hereinabove, the metal coordination compound used in the present invention provides a higher phosphorescence efficiency and a shorter phosphorescence life and allows control of its emission wavelength by appropriately modifying the substituents X1 to X8, thus being suitable as a luminescent material for EL device.

[0105]

The luminescence device having an organic layer containing the metal coordination compound is an excellent device which exhibits a high efficiency luminescence, a high luminance for a long period, and a less luminescence deterioration in energized state.

[Brief Description of the Drawings]

Figure 1 is a view showing an embodiment of a luminescence device according to the present invention.

Figure 2 is a graph showing a relationship between a Hammett's substitution constant σ and a peak (maximum) emission wavelength in the metal coordination compound of the present invention.

[Explanation of Symbols]

11: metal electrode

12: luminescence layer

13: hole-transporting layer

14: transparent electrode

15: transparent substrate

16: electron-transporting layer

17: exciton diffusion-prevention layer

【DOCUMENT NAME】 DRAWINGS

FIG. 1

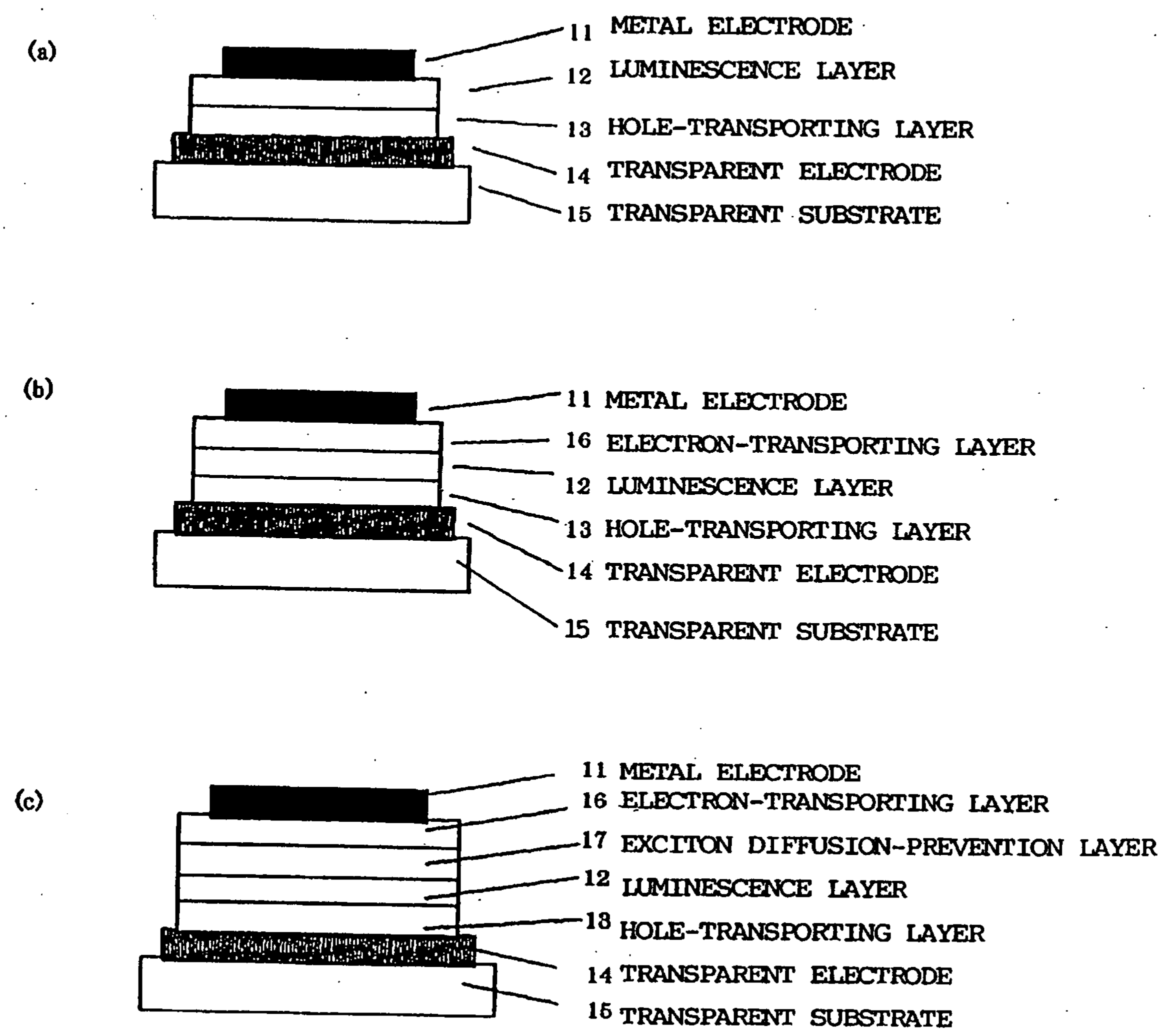
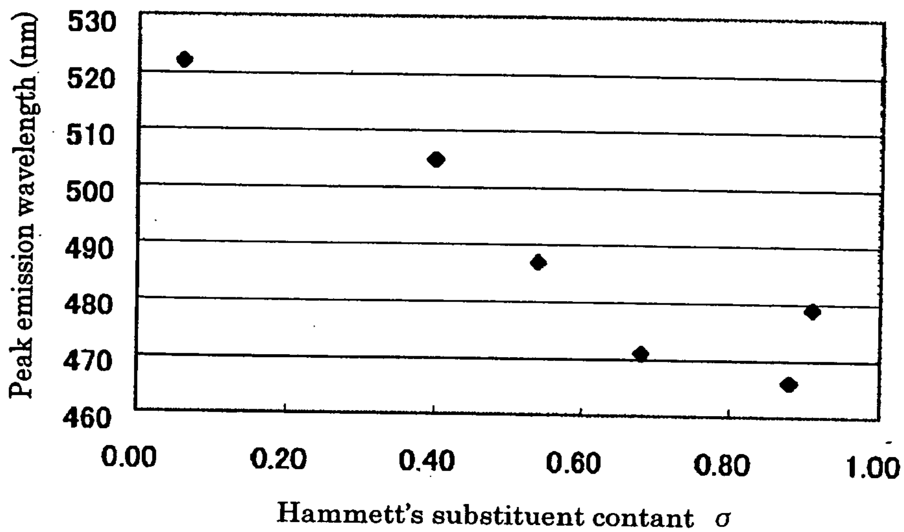


FIG. 2



Peak emission wavelength (nm)
530
520
510
500
490
480
470
460
0.00
0.20
0.40
0.60
0.80
1.00
Hammett's substituent contant σ

[Document Name]

Abstract

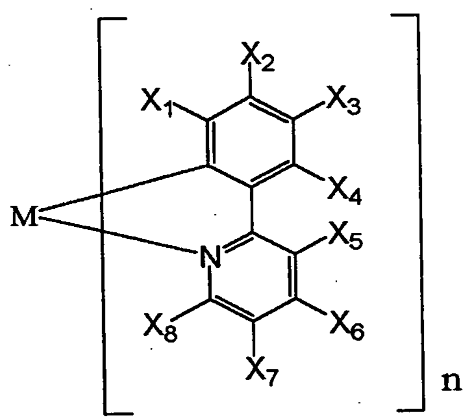
[Abstract]

[Problem]

To provide a luminescence device which produces high-efficiency luminescence, maintains a high brightness for a long period of time, and exhibits less deterioration in energized state.

[Solution Means]

A luminescence device including an organic compound layer which contains a metal coordination compound having a partial structure represented by the following formula (1):



(1).

[Selected Figure]

Figure 1